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## HEAT AND MASS TRANSFER AND PROPERTIES OF WORKING FLUIDS AND MATERIALS

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# Calculating the Caloric Properties of Dissociated Steam at High Temperatures

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**Abstract**—Lookup tables and the  $h$ - $s$ -diagram of the caloric parameters of dissociated steam are drawn up in the temperature and pressure ranges 1250–4000 K and 0.1–10 MPa for calculating the processes in thermal power installations that use dissociated steam as working fluid. Unlike the existing tables for thermophysical properties of dissociated steam, the enthalpy presented in the proposed tables has positive values in the entire range of the considered parameters. The enthalpy of mixture includes the entire energy spent in the chemical reaction of dissociation for breaking the bonds in steam molecules. The caloric parameters are determined from the reference temperature equal to 0°C adopted in the methods for calculating the parameters of non-dissociated steam, due to which it becomes possible to carry out coordinated calculations of processes in thermal power installations. Supplements to the existing tables for calculating the above-mentioned caloric parameters are developed. Dissociated steam expansion processes are calculated for the conditions with and without recombination of dissociation products at the same values of initial parameters and final pressure. It has been determined that the change of enthalpy and the work done in the expansion process without recombination are smaller than they are in the process involving recombination of dissociation products. The error of the performed calculations of caloric parameters is less than 0.1%.

**Keywords:** dissociated steam, enthalpy, entropy, formation heat, reference temperature, thermal power installations

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The progress achieved in the development of hydrogen technologies and obtaining of high-temperature steam by oxidizing hydrogen in oxygen medium generated the need to determine the thermodynamic properties, first of all, the caloric parameters of such steam. The data available in the existing tables [1] do not allow one to determine the full value of enthalpy, and the adopted reference temperature equal to 0 K is not used in thermodynamic calculations.

Steam begins to dissociate at temperatures higher than 1300 K. Hydrogen combustion and dissociation of steam are reversible chemical reactions. In our analysis, we assume that on reaching equilibrium, the chemically reacting mixture formed as a result of hydrogen combustion or dissociation of steam at the same temperatures contains the same main components: molecules of hydrogen  $H_2$ , hydroxyl  $OH$ , oxygen  $O_2$ , and atoms of hydrogen  $H$  and oxygen  $O$  [2].

The degree of dissociation depends on temperature and pressure: it increases with increasing the temperature and decreasing the pressure. At moderate pressures, dissociated steam can be regarded as a mixture of ideal chemically reacting gases. The properties of individual gases in the mixture are close to those of ideal gases; however, the mixture does not obey the laws of ideal gases; the higher the thermal effects of chemical reactions, the higher the degree to which the

properties of mixture deviate from those of ideal gases. This difference is due to changes occurring in the composition of mixture components in changing the temperature [3, 4].

The methodical principles of calculating the thermodynamic properties of thermally dissociated steam are presented in [3]. The detailed tables for the thermodynamic properties of dissociated steam in the wide ranges of temperatures from 1250 to 6000 K and pressures from 0.01 to 100 MPa with the reference temperature 0 K were calculated by N.B. Vargaftic and are given in the handbook [1]. The enthalpy of dissociated steam has negative or positive values depending on the pressure and temperature.

As dissociated steam expands in heat engines, its temperature decreases, the dissociation products recombine into  $H_2O$  molecules, and steam transforms into its usual state. So, if dissociated steam is used as working fluid in heat engines, its parameters in all states must be calculated with respect to the same reference temperature and with commensurable values of enthalpy and entropy. In engineering calculations, the thermodynamic parameters of nondissociated steam are determined with respect to the reference temperature 0°C (273.15 K). In view of this, the thermodynamic properties of dissociated steam presented in [1] can be used in thermal engineering calculations only if

supplements in the form of corrections are introduced to the presented tables for calculating the enthalpy and entropy with the reference temperature 273.15 K, and also if the enthalpy is converted into a positive quantity in the range of considered parameters.

For calculating the tables of caloric parameters and elaborating the supplements, we used the equation for the thermal effect from the reaction of thermal steam dissociation, as a result of which a chemically reacting mixture is produced. The composition of mixture was calculated using the equilibrium constants of the independent chemical reactions possible in the given thermodynamic system, and the thermal effect was calculated using the data on the enthalpy  $H(T)$  of each component contained in dissociated steam.

The absolute value of enthalpy  $H(T)$  cannot be determined experimentally or theoretically [5]. However, for solving engineering problems, it is sufficient to know the changes of thermodynamic properties in the chemical reactions, or when changes occur in the state parameters of substance, and to determine certain state of substance as the basic or standard state, from which the changes of its properties could be referred.

The Commission for Thermodynamics of the International Union for Theoretical and Applied Chemistry recommended to adopt the state of hypothetical ideal gas and the state of pure substance at a pressure of 101 325 Pa as the standard states for gases and liquids or solid substances, respectively. The thermal effects of reactions under the standard conditions are calculated taking into account the heat of product formation from simple elements at the temperatures equal to 0 or 298.15 K [5]. The change of system enthalpy in chemical reactions that occurs in changing the temperature  $T$  is determined by the thermal effect of reaction in the isobaric process, for which it is convenient to use the following calculation expression if tables of enthalpy vs. temperature are available [5]:

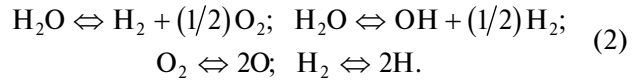
$$Q(T) = Q^0(0) + [H(T) - H(0)]_{\text{pr}} - [H(T) - H(0)]_{\text{in}}, \quad (1)$$

where  $Q^0(0)$  is the thermal effect of the chemical reaction for the adopted reference temperature  $T_0$ , which is equal to the difference between the heat of product formation and the heat of initial reacting substances; the superscript "0" denotes the standard state of substance at the pressure  $p = 101\,325$  Pa; and  $[H(T) - H(0)]_{\text{pr}}$  and  $[H(T) - H(0)]_{\text{in}}$  is the change in the enthalpies of chemical reaction products and initial substances when the temperature changes from  $T_0$  to its current value  $T$ .

In what follows, we will use the term "enthalpy" instead of the change of substance enthalpy in the temperature range from the reference point to the current temperature.

The parameters of dissociated steam are calculated for an equilibrium mixture. The equilibrium state of

mixture is determined by the equilibrium constant. For recalculating the thermodynamic parameters of dissociated steam we will use the procedure adopted in [1, 3], in which the steam dissociation processes are described by the following independent reversible reactions:



The equilibrium composition of components for these reactions is calculated from the mass action law using the equilibrium constants [3]

$$\begin{aligned} \frac{K_{\text{eq1}}}{p^{0.5}} &= \frac{x_{\text{H}_2}x_{\text{O}_2}^{0.5}}{x_{\text{H}_2\text{O}}}, & \frac{K_{\text{eq2}}}{p^{0.5}} &= \frac{x_{\text{OH}}x_{\text{H}_2}^{0.5}}{x_{\text{H}_2\text{O}}}, \\ \frac{K_{\text{eq3}}}{p} &= \frac{x_{\text{O}}^2}{x_{\text{O}_2}}, & \frac{K_{\text{eq4}}}{p} &= \frac{x_{\text{H}}^2}{x_{\text{H}_2}}, \end{aligned}$$

where  $K_{\text{eq1}}$ ,  $K_{\text{eq2}}$ ,  $K_{\text{eq3}}$ , and  $K_{\text{eq4}}$  are the equilibrium constants of the gas mixture components;  $x_{\text{H}_2}$ ,  $x_{\text{O}_2}$ ,  $x_{\text{H}_2\text{O}}$ ,  $x_{\text{OH}}$ ,  $x_{\text{O}}$ , and  $x_{\text{H}}$  are the mole fractions of the mixture components: molecules of hydrogen, oxygen, steam, and hydroxyl, and atoms of oxygen and hydrogen; and  $p$  is the pressure of gas mixture, bar (101 325 Pa).

For the possibility to solve it, this system of equations is supplemented by the equations of mass action law and balance of atoms; the latter is determined using the ratio of O and H atoms in the initial substance ( $\text{H}_2\text{O}$ ) and in the dissociation products. By solving this system, we determine the equilibrium composition of the components at the specified values of pressure  $p$  and temperature  $T$ .

In calculating the equilibrium composition of components, dissociated steam is considered in [1, 3] as a mixture of components the reality of which was taken into account by calculating their activity using the second and third virial coefficients. In the handbook [1], the dissociated steam mixture composition was determined at temperatures  $T \geq 2000$  K. Table 1 contains the calculated compositions of mixture in the temperature range 1300–1900 K.

In the steam dissociation chemical reaction, steam is the initial substance, and the components of the formed mixture are the dissociation products. The thermal effect of the reaction according to (1) and (2) can be represented in the form

$$\begin{aligned} Q_{\text{ds}}(T) &= \sum_1^3 Q_i^0(0)x_i - Q_{\text{H}_2\text{O}}^0(0)(1 - x_{\text{H}_2\text{O}}) \\ &+ \sum_1^5 H_i(T)x_i - H_{\text{H}_2\text{O}}(T)(1 - x_{\text{H}_2\text{O}}), \end{aligned} \quad (3)$$

where

$$\sum_1^3 Q_i^0(0)x_i = Q_{\text{OH}}^0(0)x_{\text{OH}} + Q_{\text{H}}^0(0)x_{\text{H}} + Q_{\text{O}}^0(0)x_{\text{O}}$$

**Table 1.** Molar composition of dissociated steam mixture components

$T, K$	$O_2$	$O$	$H_2$	$OH$	$H$	$H_2O$
<b>0.1 MPa</b>						
1400	0.00004	0.00000	0.00009	0.00001	0.00000	0.99985
1500	0.00007	0.00000	0.00017	0.00004	0.00000	0.99972
1600	0.00019	0.00000	0.00044	0.00010	0.00000	0.99927
1700	0.00043	0.00000	0.00097	0.00024	0.00001	0.99835
1800	0.00078	0.00000	0.00182	0.00055	0.00002	0.99684
1900	0.00137	0.00001	0.00330	0.00111	0.00005	0.99416
<b>1.0 MPa</b>						
1400	0.00003	0.00000	0.00005	0.00000	0.00000	0.99992
1500	0.00005	0.00000	0.00010	0.00002	0.00000	0.99984
1600	0.00010	0.00000	0.00022	0.00004	0.00000	0.99964
1700	0.00021	0.00000	0.00047	0.00011	0.00000	0.99921
1800	0.00037	0.00000	0.00087	0.00025	0.00000	0.99850
1900	0.00063	0.00000	0.00152	0.00052	0.00001	0.99731
<b>10.0 MPa</b>						
1400	0.00001	0.00000	0.00003	0.00000	0.00000	0.99995
1500	0.00003	0.00000	0.00006	0.00001	0.00000	0.99991
1600	0.00005	0.00000	0.00012	0.00002	0.00000	0.99981
1700	0.00009	0.00000	0.00020	0.00005	0.00000	0.99966
1800	0.00018	0.00000	0.00042	0.00011	0.00000	0.99929
1900	0.00030	0.00000	0.00072	0.00024	0.00000	0.99873

is the sum of the products of the standard formation heat values at the temperature 0 K  $[Q_{OH}^0(0), Q_H^0(0), Q_O^0(0)]$  of dissociation products OH, H, and O by their mole fractions;  $Q_{H_2O}^0(0)(1 - x_i)$  is the product of standard  $H_2O$  formation heat at the temperature  $T_0 = 0$  K by the mole fraction  $(1 - x_i)$  of  $H_2O$  steam converted into dissociation products;

$$\sum_{i=1}^5 H_i(T)x_i = H_{H_2}x_{H_2} + H_{O_2}x_{O_2} + H_{OH}x_{OH} + H_Hx_H + H_Ox_O$$

is the sum of the products of enthalpies by the mole fractions of mixture components  $H_2$ ,  $O_2$ ,  $OH$ ,  $H$ , and  $O$  at temperature  $T$ ; and  $H_{H_2O}(T)$  is the change of  $H_2O$  enthalpy from the reference temperature to the reaction temperature  $T$ .

In accordance with the definition adopted in [1, 3], the enthalpy of dissociated steam  $H_{mix-}$  can be a negative quantity. The equation for calculating it can be presented as part of the expression for the thermal

effect of steam dissociation reaction (3) after eliminating the terms  $[Q_{H_2O}^0(0) - H_{H_2O}(T)]$ :

$$H_{mix-} = Q_{H_2O}^0(0)x_{H_2O} + \sum_{i=1}^3 Q_i^0(0)x_i + \sum_{i=1}^5 H_i(T)x_i + H_{H_2O}(T)x_{H_2O}, \quad (4)$$

where  $Q_{H_2O}^0(0)x_{H_2O}$  is the mole fraction of the nonutilized  $H_2O$  formation heat in the steam dissociation reaction,  $H_i(T)$  is the change in the enthalpy of  $H_2O$  steam dissociation products from the reference temperature 0 K to the reaction temperature [1], and  $x_i$  is the mole fraction of the  $i$ th mixture component.

In engineering thermodynamics, the enthalpy of dissociated steam can be defined as the total amount of energy spent in an isobaric process for heating steam, for the endothermic reactions of breaking bonds in the molecules of  $H_2O$ ,  $H_2$ , and  $O_2$ , and for increasing the temperature of the produced mixture. The enthalpy calculated in this way will remain positive with the temperature varying in the entire range from 0 to  $T$ .

The equation for calculating the positive enthalpy of the chemically reacting mixture produced in the steam dissociation process is part of the expression for calculating the thermal effect from steam dissociation reaction (3) after eliminating the enthalpy  $H_{\text{H}_2\text{O}}(T)$  from it:

$$H_{\text{mix}+} = \sum_{i=1}^3 Q_i^0(0)x_i - Q_{\text{H}_2\text{O}}^0(0)(1 - x_{\text{H}_2\text{O}}) + \sum_{i=1}^5 H_i(T)x_i + H_{\text{H}_2\text{O}}(T)x_{\text{H}_2\text{O}}. \quad (5)$$

Equation (5) includes the total amount of energy spent for breaking the bonds in the  $\text{H}_2\text{O}$ ,  $\text{H}_2$ , and  $\text{O}_2$  molecules, and equation (4) includes the nonutilized heat of  $\text{H}_2\text{O}$  formation and the formation heat of dissociation products  $\text{OH}$ ,  $\text{H}$ , and  $\text{O}$ . The two first terms in equation (5) determine the expenditure of energy for breaking the bonds in molecules, and the next two terms determine the energy spent for increasing the enthalpy of dissociation products and the nonreacted  $\text{H}_2\text{O}$  steam, which is a constituent part of the mixture. For substantiating the possibility of using the tables in handbook [1] for calculating the positive enthalpies of mixture, we transform equation (5) to the form

$$H_{\text{mix}+} = -Q_{\text{H}_2\text{O}}^0(0) + Q_{\text{H}_2\text{O}}^0(0)x_{\text{H}_2\text{O}} + \sum_{i=1}^3 Q_i^0(0)x_i + \sum_{i=1}^5 H_i(T)x_i + H_{\text{H}_2\text{O}}(T)x_{\text{H}_2\text{O}}. \quad (6)$$

It follows from a comparison of equations (6) and (4) that equation (6) contains the additional term  $Q_{\text{H}_2\text{O}}^0(0)$  corresponding to the standard heat of  $\text{H}_2\text{O}$  formation, using which it is possible to calculate the positive steam enthalpy from the reference point 0 K to the reaction temperature from the tables in [1]. For calculating the dissociated steam enthalpy at the reference temperature equal to 273.15 K, we determined the values of  $\text{OH}$ ,  $\text{H}$ ,  $\text{O}$ , and  $\text{H}_2\text{O}$  formation heat at this temperature. The enthalpy of  $\text{H}_2\text{O}$  steam was calculated from the system of equations given in [6]. For temperature  $T > 2500$  K, the steam parameters were found from the tables given in [7].

The change of entropy  $\Delta S_{\text{ds}}$  in the chemical reaction with changing the reference temperature  $T_0$  is determined by the difference between the entropy values of reaction products at the current reaction temperature  $T$  and at the reference temperature  $T_0$ . For the steam dissociation reaction, the change of entropy can be expressed by the equation

$$\Delta S_{\text{mix}} = \sum_{i=1}^6 S_i(T)x_i - \sum_{i=1}^6 S_i(T_0)x_{i0}, \quad (7)$$

where  $\sum_{i=1}^6 S_i(T)x_i$  is the entropy of the mixture of components  $\text{OH}$ ,  $\text{H}_2$ ,  $\text{O}_2$ ,  $\text{H}$ ,  $\text{O}$ , and  $\text{H}_2\text{O}$ ;  $S_i(T) = S_i^0(T_0) + S_i(\Delta T)$  is the entropy of the individual component of mixture at the current temperature  $T$ ; and  $S_i(\Delta T)$  is the correction that takes into account the change of entropy when the temperature increases from  $T_0$  to the current temperature  $T$  due to nonideal properties of the components.

If the chemical reaction of dissociation does not occur at the reference temperature  $T_0$ , equation (7) becomes

$$\Delta S_{\text{mix}} = \sum_{i=1}^6 S_i(T)x_i - S_{\text{H}_2\text{O}}^0(T_0). \quad (8)$$

According to the Nernst theorem, the entropy at the temperature 0 K is equal to zero; therefore, the entropy of dissociated steam mixture with the reference temperature equal to 0 K is determined in [1]

from the expression  $S_{\text{mix}} = \sum_{i=1}^6 S_i(T)x_i$ , and with the reference temperature equal to 273.15 K it is determined from the expression

$$\Delta S_{\text{mix}} = \sum_{i=1}^6 S_i(T)x_i - S_{\text{H}_2\text{O}}(273.15), \quad (9)$$

where  $S_{\text{H}_2\text{O}}(273.15) = 63.356$  kJ/(kmol K) is the entropy of 1 kmol of water at 273.15 K [5].

All formulas presented above are related to molar quantities.

The specific values (related to 1 kg) are determined by dividing the molar quantities by the apparent molar mass of the mixture:

$$M_{\text{mix}} = \sum M_i x_i,$$

where  $M_i$  is the molecular mass of the  $i$ th component of mixture.

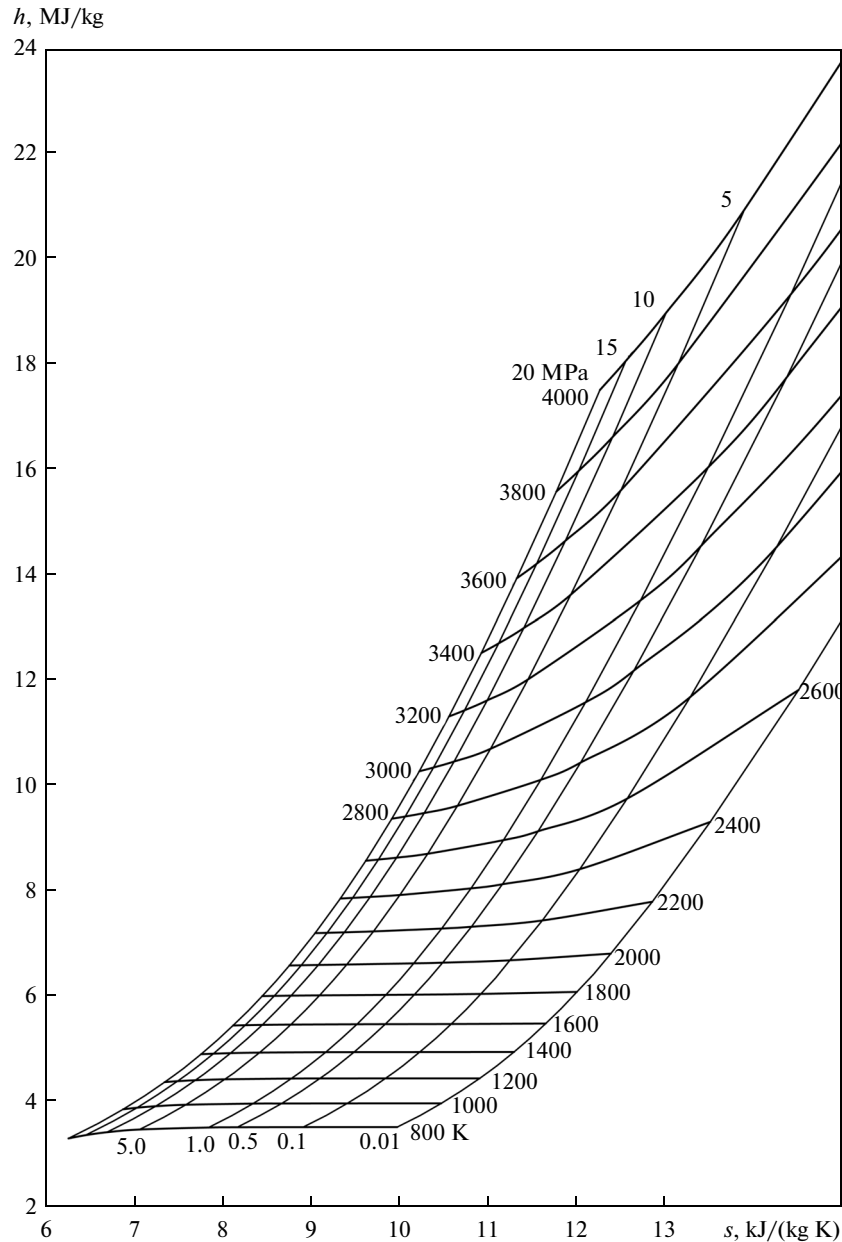
In accordance with (9), the specific entropy presented in [1] decreases with the considered change of reference temperature 273.15 K by  $3.519 \pm 0.002$  kJ/(kg K) [5].

Using equations (6) and (9) we calculated the molar values of caloric parameters from the reference temperature equal to 0°C (273.15 K) taking into account the real properties of components in the temperature range 1250–4000 K for pressures equal to 0.1, 1, 5, and 10 MPa. The calculated specific values of enthalpy and entropy are summarized in Table 2, in which the values of steam dissociation degree  $\alpha$  as functions of pressure and temperature are also given.

In our calculations of steam parameters carried out for the reference temperature equal to 0 K, the enthalpy values deviated from those presented in the handbook [1] by no more than  $\pm 5$  kJ/kg and the entropy values, by no more than 0.005 kJ/(kg K). The error with which the thermodynamic parameters were calculated

**Table 2.** Caloric parameters of dissociated steam

$T, K$	0.1 MPa			1.0 MPa			5.0 MPa			10.0 MPa		
	$s, \text{kJ}/(\text{kg K})$	$h, \text{kJ}/\text{kg}$	$a$	$s, \text{kJ}/(\text{kg K})$	$h, \text{kJ}/\text{kg}$	$a$	$s, \text{kJ}/(\text{kg K})$	$h, \text{kJ}/\text{kg}$	$a$	$s, \text{kJ}/(\text{kg K})$	$h, \text{kJ}/\text{kg}$	$a$
1250	9.937	4585	—	8.872	4582	—	9.937	4585	—	7.791	4555	—
1300	10.035	4710	—	8.97	4707	—	10.035	4710	—	7.892	4682	—
1400	10.223	4966	0.0003	9.159	4964	—	10.223	4966	0.0003	8.084	4939	—
1500	10.404	5225	0.0005	9.339	5222	—	10.404	5225	0.0005	8.267	5202	—
1600	10.577	5494	0.0008	9.511	5490	0.0008	10.577	5494	0.0008	8.441	5472	—
1700	10.747	5766	0.0014	9.678	5767	0.0013	10.747	5766	0.0014	8.607	5749	0.0004
1800	10.913	6065	0.003	9.84	6049	0.0019	10.913	6065	0.003	8.768	6031	0.0008
1900	11.08	6364	0.0058	9.998	6331	0.0025	11.08	6364	0.0058	8.923	6315	0.0014
2000	11.239	6690	0.0104	10.155	6634	0.0032	11.239	6690	0.0104	9.075	6604	0.0022
2200	11.61	7441	0.0275	10.47	7289	0.0085	11.61	7441	0.0275	9.37	7217	0.0058
2400	12.032	8405	0.062	10.8	8040	0.0189	12.032	8405	0.062	9.662	7880	0.013
2600	12.565	9732	0.1232	11.166	8944	0.0375	12.565	9732	0.1232	9.959	8616	0.0259
2800	13.277	11651	0.2197	11.589	10081	0.0679	13.277	11651	0.2197	10.272	9455	0.0469
3000	14.255	14489	0.3545	12.095	11550	0.1132	14.255	14489	0.3545	10.612	10463	0.081
3200	15.599	18670	0.5187	12.715	13471	0.1766	15.599	18670	0.5187	10.982	11578	0.1219
3400	17.394	24607	0.6878	13.481	15998	0.2596	17.394	24607	0.6878	11.4	12956	0.1792
3600	19.617	32398	0.8294	14.419	19291	0.362	19.617	32398	0.8294	11.87	14604	0.25
3800	22.025	41306	0.9224	15.551	23481	0.4794	22.025	41306	0.9224	12.403	16574	0.3328
4000	24.205	49796	0.9697	16.871	28632	0.6036	24.205	49796	0.9697	13.003	18917	0.4245

The  $hs$ -diagram of dissociated steam.

in [1] is not indicated, and in [3] it is pointed out that the obtained data are in good agreement with the results of previous calculations.

The positive enthalpy can be determined from the tables given in [1] using the correction. In accordance with equation (6), the specific enthalpy of dissociated steam for the reference temperature  $0^\circ\text{C}$  is

$$h_{\text{mix}+} = \frac{Q_{\text{H}_2\text{O}}(273)}{M_{\text{H}_2\text{O}}} + \delta h + h_{\text{mix}-}(0) = \delta h_{\text{mix}} + h_{\text{mix}-}(0), \quad (10)$$

where  $\delta h_{\text{mix}}$  is the correction to the tabular enthalpy values given in [1] for the change of formation heat

and enthalpy of mixture components in changing the reference temperature. For preliminary calculations of specific positive enthalpy at the reference temperature  $0^\circ\text{C}$  in the range of considered parameters, it is possible to take a constant value of correction independent of the dissociated steam parameters,  $\delta h_{\text{mix}} = 15\,256\text{ kJ/kg}$ . The absolute error of the correction is  $\pm 15\text{ kJ/kg}$ .

After that, we studied the effect the rate of chemical reactions has on the dissociated steam expansion process. Two cases were considered: steam expansion process with and without recombination of dissociation products. It has been found that if no chemical reac-

**Table 3.** Results from comparative calculations of chemically reacting and nonreacting mixture expansion processes

Initial parameters				Final parameters									
Dissociated steam				dissociated steam expansion with recombination of mixture components					gas mixture expansion without recombination of components				
$p$ , MPa	$T_0$ , K	$h$ , kJ/kg	$s$ , kJ/(kg K)	$p_f$ , MPa	$T_0$ , K	$h$ , kJ/kg	$\Delta h$ , kJ/kg	$L$ , kJ/kg	$p_f$ , MPa	$T_0$ , K	$h$ , kJ/kg	$\Delta h$ , kJ/kg	$L$ , kJ/kg
10	3600	14638	11.867	0.1	2321	8064	6574	6574	0.1	1658	8602	6036	6036
1	2600	8983	11.166	0.1	1954	6552	2431	2431	0.1	1805	6656	2327	2327

tion occurs during the expansion process to the preset pressure, the work  $L$  is smaller than it is when the dissociation products undergo recombination. This is attributed to the fact that no energy is released in the gas expansion process if there is no chemical reaction. The calculation results are given in Table 3.

It follows from the calculation results given in Table 3 that in studying the processes in heat engines that use dissociated steam as working fluid, it is necessary to take into account the energy of chemical reaction occurring in the reacting mixture, the value of which depends on the steam parameters, and, as it follows from Table 3, the chemical energy increases with the temperature.

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